OFFICE OF NAVAL RESEARCH
DEPARTMENT OF THE NAVY
CONTRACT N00014-67-0094-0009

TENSILE STRENGTH OF LIQUIDS

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PASADENA, CALIFORNIA

REPORT NO. 85-47

APRIL 1969

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ABSTRACT

The theory of the tensile strength of a pure liquid is developed and it is shown that it predicts much larger tensile strengths than are observed. This theory is modified and extended under the supposition that liquids usually contain nuclei which are here taken to be solid particles. It is shown that the theory leads to more moderate predictions of tensile strength provided the solid particles are not wetted by the liquid. It is also shown that Brownian motion will serve as the mechanism whereby solid particles can remain in suspension in liquids.

Introduction

A central problem in cavitation and boiling is how macroscopic vapor cavities can form when moderate tensions are applied to a liquid. The theory of the tensile strength of pure liquids predicts that a vapor cavity will form only when the liquid is under extremely large tensions; as an equivalent effect the theory also predicts that vapor bubbles appear in boiling only when the liquid has very large superheats. Since these large tensile strengths and superheats are not observed, the idea of nuclei has been introduced. These nuclei are in some sense holes in the liquid which are already beyond molecular dimensions and which may therefore grow into macroscopic bubbles under moderate liquid tensions. Gas bubbles of small size can not themselves serve as nuclei because they will dissolve fairly rapidly except under a very special condition of supersaturation of dissolved gas[1] [1]. Further in this condition of supersaturation, the gas bubble can not serve as a persistent nucleus since the bubble is unstable relative both to a slight variation in the bubble radius as well as to a slight variation in the dissolved gas concentration. In order to avoid this stability difficulty it has been suggested [2] that a gas bubble in a liquid acquires an organic, possibly protein, skin over its boundary. Such a skin might have two effects: first, the skin might give some structural integrity to the bubble and second, it might inhibit diffusion of gas out of the bubble into the liquid. The notion of bubbles with such skins is untenable for several reasons. Chemical reagants which in small amounts strongly affect organic materials including proteins, do not have a

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[1] Numbers in brackets designate References at the end of the paper.

noticeable effect on the observed tensile strength of liquids. It is also observed that bubbles grow quite readily by rectified diffusion in oscillating pressure fields so that there is no evidence for inhibition of diffusion at bubble boundaries.

The emphasis on gas bubbles as nuclei and on effect of dissolved gases arises presumably from the large body of experimental information based on the measurements of the tensile strength of liquids by means of oscillating pressure fields. The phenomenon of rectified diffusion of dissolved gas into bubbles is decisive for bubble growth in these sonic or ultrasonic observations. Of more fundamental importance for the theoretical tensile strength are measurements made with static tensions or even with transient tensions. With such pressure fields, dissolved gas does not play a significant role.

We shall adopt the view here that the nuclei in a liquid are solid particles, and our objective will be to determine if the moderate tensile strengths which are observed can be predicted with this picture on a reasonable, quantitative, physical basis. We are concerned with "moderately" clean liquids. Dirty liquids have very low tensile strengths or no tensile strength at all, and these liquids usually have visible impurities or are in contaminated containers. Such liquids are not ordinarily the subject of serious laboratory experimentation. By a moderately clean liquid, on the other hand, we shall mean a reasonably pure liquid. Such a liquid has a stable value of tensile strength which is reproducible, and extraordinary purification treatment of both the liquid and its container as well as isolation from the atmosphere is required if a significant increase in tensile strength is

to be obtained. Even carefully purified liquid samples, however, have measured tensile strengths which are significantly lower than the theoretically predicted values. If the tensile strengths of such liquids are to be understood in terms of solid nuclei, one must explain the stability and reproducibility of the observations. If the observed tensile strengths require nuclei which are too large, such nuclei would not only be easily visible but would also settle out of suspension in a quiescent liquid fairly quickly. It is therefore necessary to consider the stability of the distribution of solid nuclei under the action of gravity.

The following analysis will begin with a presentation of the theory of the ideal tensile strength of a pure liquid. While the results of the theory for a pure liquid free of nuclei are not entirely new, it is hoped that the analysis given here has the advantages of brevity and clarity. It will also be useful to have this theory for the pure liquid available in a form which allows easy extension to liquids with nuclei. This extension of the calculation to liquids with nuclei, or "real liquids", will bring out those properties of nuclei which are essential to explain experiment. Finally, the stability under gravity of the suspension of solid nuclei in liquids will be considered.

Tensile Strength of a Pure Liquid

We wish to calculate the probability of the transition of a pure homogeneous liquid into a liquid with a vapor bubble of a radius R. We make the calculation of this probability definite by supposing that it takes place isothermally and with the minimum increase in free

energy, that is, reversibly. We may then describe the system by the canonical distribution of statistical mechanics. If U is the internal energy of a system in its most stable state, U is also its mean internal energy and, if $F(U)$ is the free energy of the most stable state, then the probability of a fluctuation occurring to a state with internal energy in the range E to $E + dE$ is

$$P(E)dE = e^{\alpha[F(U) - E]}\omega(E)dE \qquad (1)$$

where $\alpha = 1/kT$ with k the Boltzmann constant ($k = 1.38 \times 10^{-16}$ ergs/°K), and T is the absolute temperature. The weighting function $\omega(E)$ is related to the entropy $S(E)$ of the system when the system has internal energy E:

$$\omega(E) = e^{S(E)/k}$$

so that we see that the probability $P(E)$ may be written in the form

$$P(E) = e^{F(U)/kT} \quad e^{-[E-TS(E)]/kT} \quad .$$

Since the free energy of the system with entropy $S(E)$ and internal energy E is

$$F(E) = E - TS(E) \quad ,$$

we may write

$$P(E) = e^{[F(U)-F(E)]/kT} = e^{-\Delta F/kT} \quad . \qquad (2)$$

The probability of a fluctuation at temperature T from the state U to the state E is related in this exponential manner to the increase ΔF in the free energy in going from the state U to the state E. While we may wish to replace the variable E by the radius R of the vapor bubble which appears in the metastable state, the factor so

introduced is not important since the probability P is dominated by the exponential given in Eq. (2).

The free energy $F(U)$ of the uniform liquid and the free energy $F(E)$ of the liquid with a vapor bubble may be determined in the following way. Let us suppose that the liquid in the initial stable state occupies the total volume $V + V'$ and that the pressure in the liquid is p. In the metastable state we suppose that the liquid volume is V and that the vapor bubble volume is V' at pressure p'. Further, we have initially that all the molecules, N in number, have the liquid chemical potential μ so that the initial value of the thermodynamic potential is

$$\Phi(U) = N\mu \quad .$$

In the metastable state N' molecules have passed into the vapor state with chemical potential μ' so that

$$\Phi(E) = N'\mu' + (N - N')\mu \quad .$$

We now use the general relation between the thermodynamic potential and the free energy which says that $F = \Phi$ - (pressure) $\times$ (volume). It follows that

$$F(U) = N\mu - p(V + V') \tag{3}$$

and

$$F(E) = (N - N')\mu + N'\mu' - pV - p'V' + \sigma\Sigma \quad . \tag{4}$$

The last term in Eq. (4) is the surface free energy corresponding to the surface area Σ of the interface between the liquid and vapor phases. The corresponding surface free energy is $\sigma\Sigma$ where σ is

the surface tension constant. In evaluating the free energy increment in the fluctuation, we need only to recall that an equilibrium between the liquid and its vapor at a temperature T means that $\mu' = \mu$. Then

$$- \Delta F = F(U) - F(E) = (p' - p)V' - \sigma\Sigma \quad . \tag{5}$$

The vapor cavity which appears will be quite small and surface tension forces will be much more important than gravity so that the vapor bubble will be spherical. Then the condition for mechanical equilibrium is

$$p' - p = \tau = 2\sigma/R \quad , \tag{6}$$

and $V' = 4\pi R^3/3, \Sigma = 4\pi R^2\sigma$. The free energy change is easily evaluated in this case and gives for the probability of appearance of a vapor bubble of radius R

$$P = e^{-4\pi R^2\sigma/(3kT)} \quad . \tag{7}$$

The fact that the free energy increase associated with the formation of the bubble is 1/3 the surface free energy was known to Gibbs [3]. Figure 1 gives a graph of the probability factor P as a function of R for a liquid with $\sigma = 72$ dynes/cm (water at 27° C) and for a liquid with $\sigma = 59$ dynes/cm (water at 100° C). It is evident that P becomes extremely small as soon as R exceeds molecular dimensions. For $R = 10^{-7}$ cm, $\sigma = 72$ dynes/cm, $\ell n\, P = -72.8$ and the corresponding tension is 1,440 atm. (cf. Fig. 2). For the same R, $\ell n\, P = -48$ when $\sigma = 59$ dynes/cm and the corresponding tension is approximately 1200 atm. While the tensions in the liquid go down with increasing R as shown in Fig. 2, the probability of formation of such bubbles is essentially zero. It is only when the bubble is near molecular

dimensions ($\sim 10^{-8}$ cm) that the probability of formation has an appreciable value.

The exponential factor in the probability distribution given in Eq. (7) is the dominant factor. It is of interest, however, to verify this fact by writing the probability distribution in the form of $P(R)dR$ in place of the form $P(E)dE$ given in Eq. (1). The exponential factor is already expressed in this form in Eq. (7) and all that remains is to relate dE to dR, a step that is easily made. We have the general thermodynamic relation $E = F + TS$, where E is the internal energy, F is the free energy, T is the absolute temperature and S is the entropy. These thermodynamic functions have a surface part which will be distinguished by the subscript Σ and a volume part denoted by the subscript V. We have the surface free energy $F_\Sigma = \sigma\Sigma$, and we have observed that for the spherical vapor cavity $F_V = -(2/3)F_\Sigma$. The surface entropy is $S_\Sigma = -\Sigma\, d\sigma/dT$. Thus, for an isothermal increment in the bubble volume dV' with an associated increment in interfacial area $d\Sigma$ we have

$$dE = dF_V + dF_\Sigma + T\,dS_\Sigma + T\,dS_V \ .$$

The last term gives the isothermal energy increase due to latent heat. We have

$$T dS_V = L\rho' dV'$$

where L is the latent heat of evaporation for the vapor which has density ρ'. We have, therefore

$$dE = \left\{\frac{1}{3}\sigma - T\frac{d\sigma}{dT} + L\rho'\frac{dV'}{d\Sigma}\right\} d\Sigma$$

For the spherical cavity considered here $dV'/d\Sigma = R/2$ and $d\Sigma = 8\pi R dR$ so that our probability distribution is

$$P(R)dR = 8\pi\left\{\frac{1}{3}\sigma - T\frac{d\sigma}{dT} + L\rho'\frac{R}{2}\right\}Re^{-4\pi R^2/(3kT)}dR \quad . \tag{8}$$

Equation (8) verifies the previous statement that the probability distribution is overwhelmingly dominated by the exponential factor.

Tensile Strength of Liquids with Nuclei

We now calculate the probability of formation of a vapor cavity about a nucleus in a liquid. We take the nucleus to be a rigid solid which for definiteness and simplicity is a sphere of radius R_o; we denote the volume of this sphere by $V_o = 4\pi R_o^3/3$ and its surface area by $\Sigma_o = 4\pi R_o^2$. The vapor cavity which forms about this solid particle will be spherical since, as before, gravity effects are unimportant for small cavities compared with surface tension effects. The inner boundary of the vapor cavity is the solid nucleus and the outer boundary has a radius R (cf. Fig. 3). We have $\Sigma = 4\pi R^2$ for the area of the vapor-liquid interface and write $V' = 4\pi R^3/3$.

For the stable state of the system in which there is no vapor cavity we take the pressure in the liquid to be p and the liquid volume to be $V + V' - V_o$. If the surface tension constant of the liquid-solid nucleus interface is $\sigma_{\ell n}$, the pressure at the nucleus surface is $p + 2\sigma_{\ell n}/R_o$ and the surface free energy of this interface is $\sigma_{\ell n}\Sigma_o$. The free energy of this stable system is

$$F(U) = N\mu + \sigma_{\ell n}\Sigma_o - p(V+V'-V_o) - \left(p + \frac{2\sigma_{\ell n}}{R_o}\right)V_o \quad . \tag{9}$$

In the metastable state in which we have N' vapor molecules in the volume $V'-V_o$ we have a surface free energy from the liquid-vapor interface given by $\sigma_{\ell v}\Sigma$, where $\sigma_{\ell v}$ is the surface tension constant for the liquid-vapor interface, and we also have a surface free energy at the vapor-solid nucleus interface given by $\sigma_{vn}\Sigma_o$, where σ_{vn} is the surface tension constant for the vapor-solid interface. The pressure in the liquid volume V is p, the pressure in the vapor volume $V'-V_o$ is $p' = p + 2\sigma_{\ell v}/R$, and the pressure in the volume of the solid, V_o, is $p' + 2\sigma_{vn}/R_o = p + 2\sigma_{\ell v}/R + 2\sigma_{vn}/R_o$. The free energy of the state with the vapor cavity of radius R about a nucleus of radius R_o is, therefore

$$F(E) = (N-N')\mu + N'\mu' + \sigma_{\ell v}\Sigma + \sigma_{vn}\Sigma_o - pV - \left(p + \frac{2\sigma_{\ell v}}{R}\right)(V'-V_o) - \left(p + \frac{2\sigma_{\ell v}}{R} + \frac{2\sigma_{vn}}{R_o}\right)V_o . \tag{10}$$

We keep in mind that $\mu' = \mu$, and we get for the free energy difference between the state without the bubble and the state with the bubble

$$F(U) - F(E) = \sigma_{\ell v}\left(\frac{2}{R}V' - \Sigma\right) + \sigma_{vn}\left(\frac{2}{R_o}V_o - \Sigma_o\right) - \sigma_{\ell n}\left(\frac{2}{R_o}V_o - \Sigma_o\right) .$$

It follows that

$$-\Delta F = F(U) - F(E) = -\frac{4\pi R^2}{3}\sigma_{\ell v} - \frac{4\pi R_o^2}{3}(\sigma_{vn} - \sigma_{\ell n}) . \tag{11}$$

The probability of formation of the vapor bubble of radius R about the nucleus of radius R_o is again proportional to

$$P = e^{-\Delta F/kT} . \tag{12}$$

The last term in Eq. (11) shows the dependence of our probability

function on the properties of the vapor-liquid-solid interface. The general equilibrium condition for such an interface (cf. Fig. 4) determines the angle of contact for the configuration shown which is given by

$$\cos\theta = \frac{\sigma_{vn} - \sigma_{\ell n}}{\sigma_{\ell v}} \tag{13}$$

The solid is said to be wetted by the liquid when θ is an acute angle so that the wetting condition is

$$\sigma_{vn} > \sigma_{\ell n} \tag{14}$$

We have a general result for the wetted case: the last term in Eq. (11) will then have the same sign as the first term so that the probability of formation of a bubble of radius R is <u>decreased</u> by the presence of a solid nucleus which is wetted by the liquid. It follows that the liquid will rupture under a tension at a location other than at the nucleus. Such a result is to be expected since one may think of a wetted solid as having a greater adhesion for the liquid than the cohesion of the liquid for itself.

The situation is quite different, of course, for the case of a nucleus which is not wetted by the liquid. Then $\sigma_{\ell n} > \sigma_{vn}$ which means for the general configuration shown in Fig. 4 that the angle of contact is obtuse. It is to be expected that $\sigma_{\ell n}$ may exceed σ_{vn} by varying amounts for different solids, but it is convenient for the purpose of specific calculations to suppose that $\sigma_{\ell v}$ exceeds $(\sigma_{\ell n} - \sigma_{vn})$ by a small amount; i.e., $\theta \simeq \pi$. For such a vapor-liquid-solid combination we may approximate Eq. (11)

$$\Delta F = \frac{4\pi R^2}{3}\,\sigma_{\ell v}\left[1 - \frac{R_o^2}{R^2}\right] . \tag{15}$$

The corresponding probability behavior is shown in Fig. 5 where the value of $\sigma_{\ell v}$ for water at 27°C has been used.

It is evident from Eq. (15) that the presence of a nucleus of non-wetted solid with radius R_o gives a probability which is essentially unity for the formation of a vapor bubble of this radius, and the corresponding tension under which a vapor bubble will form is $2\sigma/R_o$. If we consider for given nucleus size R_o the probability of vapor cavity formation under a tension less than $2\sigma/R_o$, we see that this probability is very small for nuclei of size $R_o = 10^{-6}$ cm or greater. This behavior is illustrated in Fig. 5 where it is shown that the probability of formation of a bubble of radius R about a nucleus of radius R_o decreases rapidly as soon as R is greater than R_o. The corresponding tension is of course $2\sigma/R$. The only situation in which this rapid decrease in probability does not occur as R/R_o increases beyond unity is for values of R_o already so small ($\sim 10^{-7}$ cm, or less) that the corresponding tensions are extremely large. The conclusion is evident that the only possibility for observing moderate tensile strengths in a liquid comes from the presence of non-wettable nuclei of sufficiently large size.

It must be pointed out that in a quiescent liquid the solid nuclei tend to settle out of suspension in a liquid. If we consider a solid particle of volume V and density ρ in a liquid of density ρ_o, then the gravitational potential energy is

$$\Phi = Vg(\rho - \rho_o)z$$

where z is the coordinate of the particle taken positive in the upward direction. The mechanism whereby particles of greater density

than the liquid remain in suspension is familiar as a Brownian motion and the probability that a particle may be found at a height z above a reference height z_o is [4]

$$P(z) = e^{-\frac{Vg(\rho-\rho_o)(z-z_o)}{kT}} \quad . \tag{16}$$

Most solid, or dust, particles which would be encountered in liquids exposed to the atmosphere would have densities in the range from 2 to 3 gm/cm^3. For spherical particles, Eq. (16) shows that a solid particle of radius $R_o = 10^{-6}$cm has a reasonable probability of remaining in suspension. On the other hand when the radius of the particle is $R_o = 5 \times 10^{-6}$cm this probability has decreased to roughly e^{-20} so that a very large population of such particles would be required for significant effect. If the liquid, however, has any appreciable transport velocity, we can expect that even particles as large as 5×10^{-6}cm will be fairly uniformly distributed since the settling velocity of such particles is so small compared to ordinary flow velocities.

Conclusions

We may give a qualitative summary of our results. A pure liquid without nuclei will have a tensile strength of thousands of atmospheres and the vapor cavities which form under the applied tension will be of the order of molecular dimensions*. A liquid with non-wetted solid particles as nuclei will show much lower tensile

*We leave out of consideration here nuclei which are formed in liquids by cosmic rays or other ionizing radiation.

strengths; in quiescent water, for example, nuclei with dimensions of the order of 10^{-6}cm will remain in suspension and will give a vapor cavity of this initial size under tensions of the order of a hundred atmospheres. If the liquid has a macroscopic flow, nuclei larger than 10^{-6}cm may remain in the body of the liquid for long times so that a tensile strengh of the order of tens of atmospheres may be expected.

A remark may be made regarding the effects of dissolved gases on the tensile strength. Two effects are to be expected. First, adsorbed films of gas may form on the surfaces of the nuclei. A significant change, however, in the effective size of the nuclei is not to be expected since the thickness of such films cannot exceed the range of long range molecular forces. The gas films, therefore, should not exceed 10^{-7}cm in thickness, and we have seen that only nuclei of 10^{-6}cm, or larger, are significant for moderate tensile strengths. A second effect of adsorbed gas films is of more interest since such films generally give a reduction in the surface tension. It is not to be expected that such a reduction is by an order of magnitude, but it is sufficient to give an observable decrease in tensile strength, a decrease which is difficult to estimate in general since it is specific for each solid-gas combination.

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2. F. E. Fox and K. F. Herzfeld, "Gas Bubbles with Organic Skin as Nuclei", J. of the Acoust. Soc., Vol. 26 (Number 6), pp. 984 - 989 (1954).

3. For different methods of arriving at results similar to Eq. (7) see L. Landau and E. Lifshitz, Statistical Physics (Addison -Wesley, 1958) §147, or M. Volmer, Kinetik der Phasenbildung (Theodor Steinkopff, 1936) pp. 97 - 100.

4. See, for example, A. Einstein, The Theory of the Brownian Movement (Dover Publication, Inc., 1956), pp. 24 - 27.

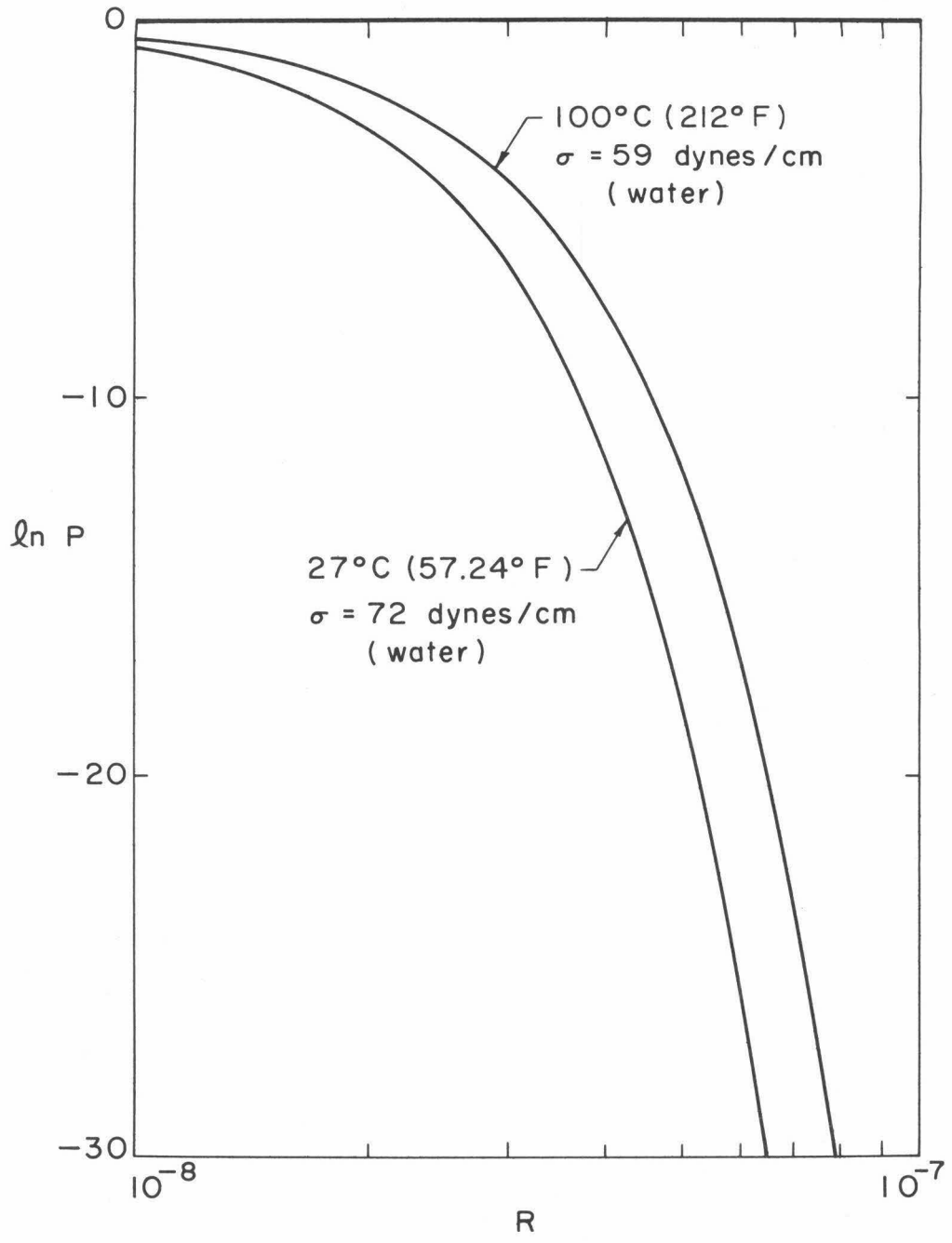


Figure 1. The natural logarithm of the probability of formation of a vapor bubble of radius R is shown for the surface tension value σ = 72 dynes/cm which is the value for water at 27°C, and for σ = 59 dynes/cm which is the value at 100°C.

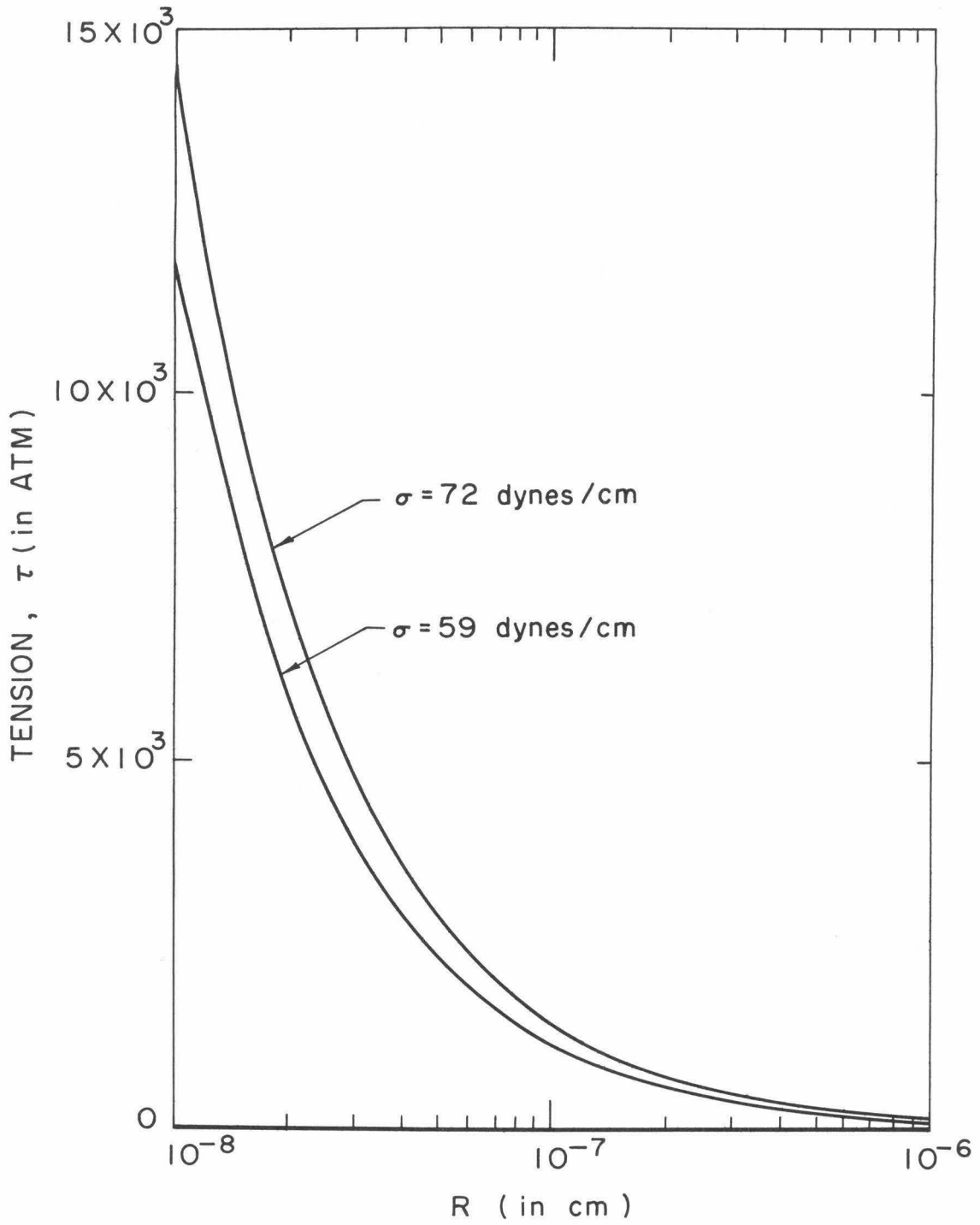


Figure 2. The tension in the liquid, τ, required to form a bubble of radius R is shown for the surface tension values for water at 27°C (72 dynes/cm) and at 100°C (59 dynes/cm).

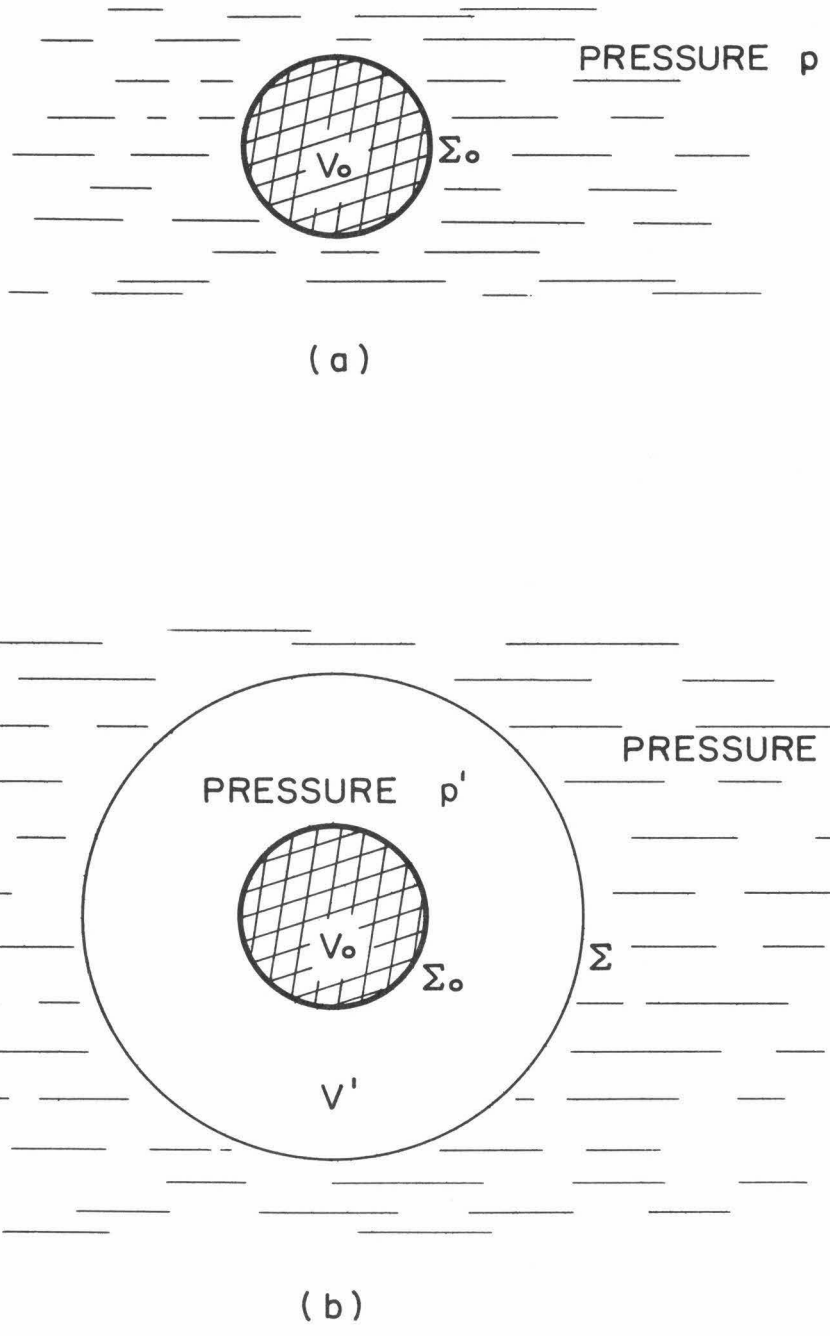
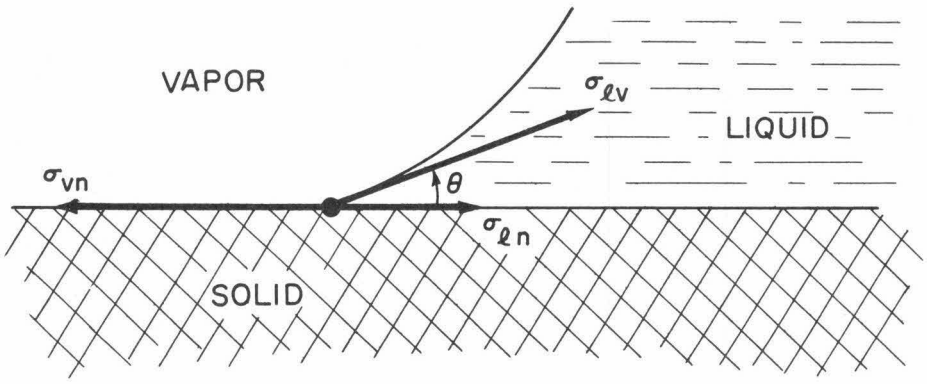
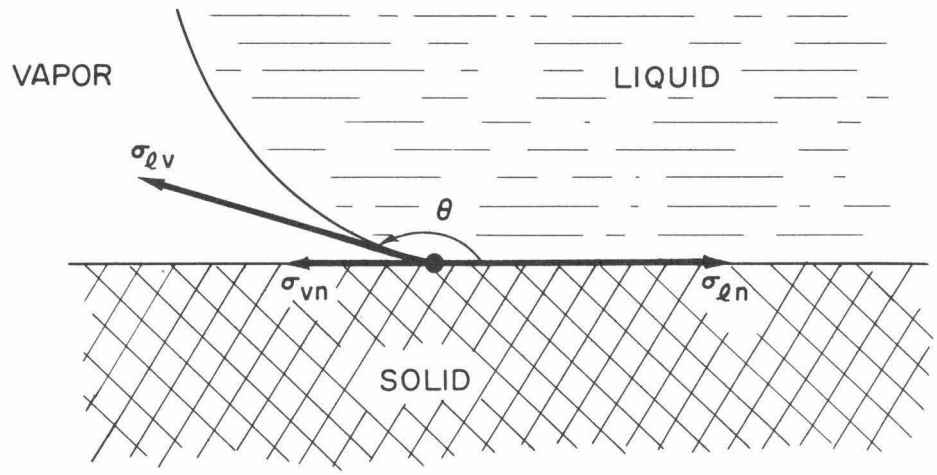


Figure 3. Figure 3a shows the geometry of a nucleus with volume V_o, surface area Σ_o, before the formation of the vapor cavity which is drawn in Figure 3b. The vapor cavity has radius R, surface area Σ and volume $V' - V_o$.



(a) SOLID WETTED BY LIQUID. $\sigma_{vn} > \sigma_{\ell n}$.



(b) SOLID NOT WETTED BY LIQUID. $\sigma_{\ell n} > \sigma_{vn}$.

Figure 4. The figure shows the general configuration for equilibrium at the contact of a liquid with a solid in the presence of the vapor phase. The acute angle (a) is characteristic of the solid wetted by the liquid and the obtuse angle (b) is characteristic for a non-wetted solid.

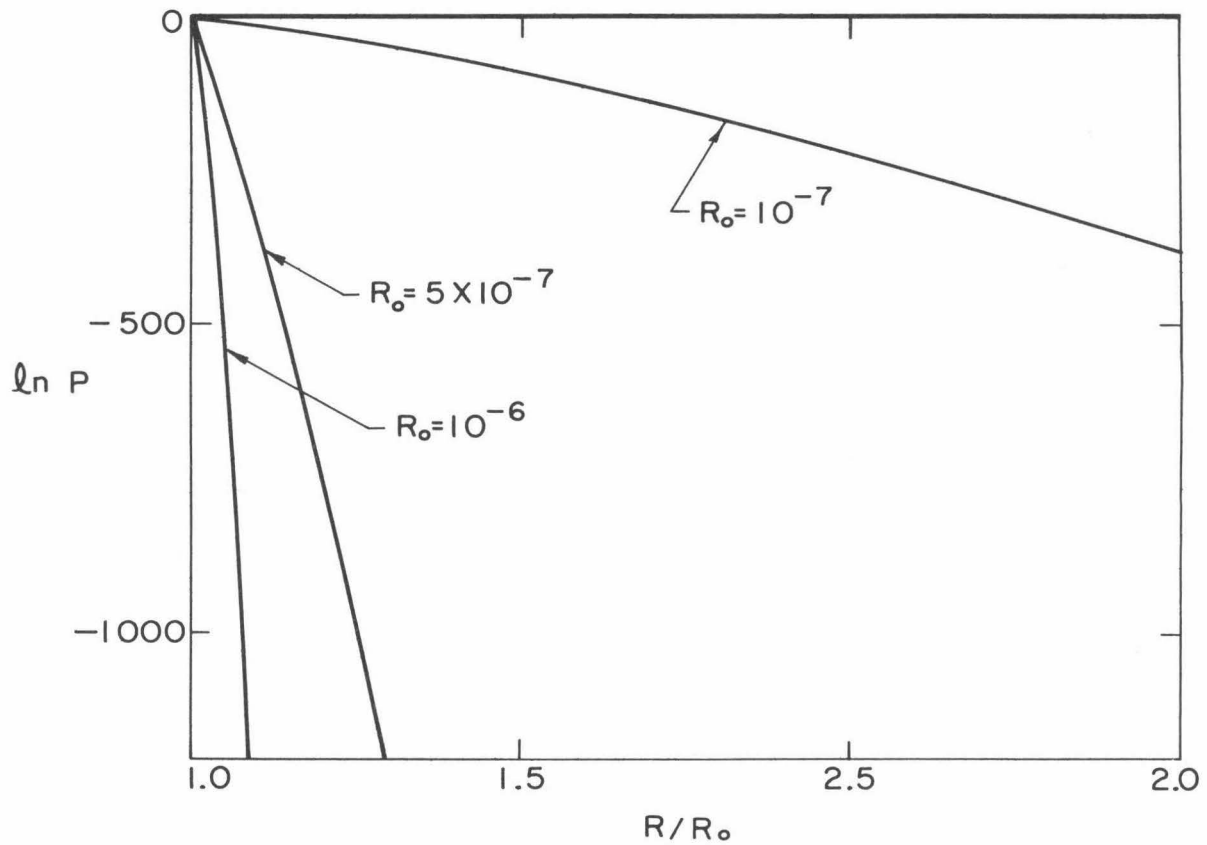


Figure 5. The natural logarithm of the probability of formation of a bubble of radius R about a nucleus of radius R_o is shown as a function of the ratio R/R_o. The surface tension value used is 72 dynes/cm (water at 27° C) and it is supposed that the solid is not wetted by the liquid with a contact angle $\cong \pi$.

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1. ORIGINATING ACTIVITY (Corporate author)	2a. REPORT SECURITY CLASSIFICATION
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	2b. GROUP Not applicable

3. REPORT TITLE

TENSILE STRENGTH OF LIQUIDS

4. DESCRIPTIVE NOTES (Type of report and inclusive dates)

Technical Report

5. AUTHOR(S) (Last name, first name, initial)

Plesset, Milton S.

6. REPORT DATE	7a. TOTAL NO. OF PAGES	7b. NO. OF REFS
April 1969	14	4

8a. CONTRACT OR GRANT NO.	9a. ORIGINATOR'S REPORT NUMBER(S)
N00014-67-0094-0009 b. PROJECT NO.	Report No. 85-47
c.	9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)
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The theory of the tensile strength of a pure liquid is developed and it is shown that it predicts much larger tensile strengths than are observed. This theory is modified and extended under the supposition that liquids usually contain nuclei which are here taken to be solid particles. It is shown that the theory leads to more moderate predictions of tensile strength provided the solid particles are not wetted by the liquid. It is also shown that Brownian motion will serve as the mechanism whereby solid particles can remain in suspension in liquids.

DD FORM 1 JAN 64 1473

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